

## **Project 94639**

### **Millimeter-Wave High Level and Low Activity Waste Glass Research**

**Woskov, Paul P.**

Massachusetts Institute of Technology

#### **RESULTS TO DATE: Research Objective**

The primary objectives of the current research is to develop on-line sensors for characterizing molten glass in high-level and low-activity waste glass melters using millimeter-wave (MMW) technology and to use this technology to do novel research of melt dynamics. Existing and planned waste glass melters lack sophisticated diagnostics due to the hot, corrosive, and radioactive melter environments. Without process control diagnostics the Defense Waste Processing Facility (DWPF) and the Waste Treatment Plant (WTP) under construction at Hanford operate by a feed forward process control scheme that relies on predictive models with large uncertainties. This scheme severely limits production throughput and waste loading. Also operations at DWPF have shown susceptibility to anomalies such as foaming and combustion gas build up, which can seriously disrupt operations. Future waste chemistries will be even more challenging. The scientific goals of this project are to develop new reliable on-line monitoring capability for important glass process parameters such as temperature profiles, emissivity, density, viscosity, and other characteristics using the unique advantages of millimeter-wave electromagnetic radiation. Once successfully developed and implemented, significant cost savings would be realized in melter operations by increasing production through put, reduced storage volumes (through higher waste loading), and reduced risks (prevention or mitigation of anomalies).

#### **Research Progress and Implications**

In the past year, as of September 2005 three research tasks were accomplished; 1) a new dual MMW receiver instrument configuration was experimented with for improved emissivity measurement, 2) laboratory studies were carried out of salt coming out of solution with glass, and 3) a MMW receiver was fabricated for the Aiken County Technology Lab (ACTL) at the Savannah River National Laboratory (SRNL) and installed on the slurry fed melt rate furnace (SMRF). These accomplishments address three project goals, respectively; 1) develop advanced MMW instrumentation for melter research and operations, in particular improved on line emissivity measurement capability may make possible monitoring small phase changes in the state of the melt such as liquidus and redox, 2) obtain more information on salt dynamics in melts which is a key concern to future vitrification operations, and 3) establish MMW instrumentation capabilities at SRNL to accelerate research on melt dynamics.

#### **1. Dual Receiver Experiments**

Previous work established the capability of detecting the emissivity of the melt glass surface by returning a part of the thermal emission to the hot surface to be reflected back to the receiver [1]. The magnitude of this reflection relative to the thermal signal without a return reflection provides a measure of the emissivity (the higher the reflectivity the lower the emissivity). In the previous work this measurement was accomplished with one receiver by periodically using a mirror to send back a part of the thermal emission via a beamsplitter located in front of the receiver. This works reasonably well when the melt temperature/properties are not changing (steady state), but is difficult under varying, transient conditions. Two receivers were set up in the past year to improve on this measurement, one with a thermal reflection and the second without, to simultaneously measure these two signals and obtain an instantaneous measure of their ratio, and consequently the emissivity. In this way small changes during transient conditions could be detected and provide new insights into the melt dynamics of the vitrification process. The dual receiver setup was implemented and experiments started. Both receivers view the sample through the same waveguide into the furnace. Initial measurements indicate that the waveguide transmission losses are an

important effect that limits the TRR enhancement of the signal in one receiver. Measurements and analysis are currently in progress to determine the effectiveness of this approach for emissivity measurements.

## 2. Salt Dynamics Studies

The melt dynamics of glass containing salt, previously observed in an engineering scale melter [2], were further studied in a laboratory furnace at MIT. The laboratory studies focused on observing salt coming out of solution from a glass frit that was previously melted with added salt content. This differed from the initial melter measurements where pure salt was added to the melt surface after a melt pool was established. Measurements with borosilicate glass S22-18 containing about 2% by weight of sodium sulfate ( $\text{NaSO}_4$ ) were carried out. The furnace temperature reached a maximum of about 1400 degree C during these measurements. Salt coming out of solution was observed as high frequency noise structure in the MMW signal. These studies are continuing and will provide valuable insights into melter salt dynamics that could be used in the future to monitor and control salt layer formation in melters.

## 3. MMW Measurements at ACTL

A Millimeter Wave Measurement System was installed and tested at the Aiken County Technical Lab (ACTL) of the Savannah River National Laboratory in Aiken, SC [3]. The system consisted of a 137 GHz heterodyne receiver box, a 4-port waveguide beam splitter block for TRR measurements, a Teflon window in a horizontal waveguide run to allow waveguide pressurization for viscosity measurements, and a vertical mullite waveguide compatible with temperatures to 1500 degree C. This new installation will allow development of MMW on-line sensor capabilities with existing melter systems, such as the Slurry fed Melt Rate Furnace. The ACTL Millimeter Wave Guide System is being setup to be portable to allow the testing of multiple melter systems. In particular, ACTL is in the process of developing and building a larger Slurry Melt Rate Furnace to allow studies of salt layer and cold cap formation for the Defense Waste Processing Facility at the Savannah River Site. This new melter installation will be integrated with the activities of the MWG at ACTL for salt layer and cold cap formation sensor studies. By leveraging the DWPF EM-50 and Office of Science DOE programs, overall research and development activities can be maximized while minimizing costs. Planned Activities

Design and testing will continue of millimeter-wave monitoring instrumentation, methodologies, and melt measurements to improve currently demonstrated molten glass parameter measurements and to develop new capabilities. This work will include analytical modeling advancements and instrument hardware and software configuration improvements to increase capabilities and ease of use. Among the areas where more research development is required are emissivity measurements, density determinations, improved viscosity modeling, and surface curvature and fluctuation effects. New diagnostic capabilities are also needed for liquidus, redox, and plenum gas temperature. Additional research would also help establish a record of operation and a database for gaining acceptance of this technology by the vitrification production facilities.

Specific tasks include: i) Develop and test advanced millimeter wave instrumentation for advanced measurements ii) Establish and exploit millimeter wave capabilities at PNNL and SRNL for melt pool dynamics measurements important to vitrification operations iii) Review and update glass matrix tested to insure all waste glass composition ranges have been considered iv) Research foaming and molten salt dynamics v) Plan integrated approach to millimeter wave real-time on-line diagnostics

**DELIVERABLES:** 1. P.P. Woskov and S. K. Sundaram, "Thermal return reflection method for resolving emissivity and temperature in radiometric methods", J. Appl. Phys., vol. 92, 6302-6310, 2002. 2. P. P. Woskov, S. K. Sundaram, W. E. Daniel, Jr., D. Miller, "Molten salt dynamics on glass melt using millimeter-wave emissivity measurements", J. of Non Crystalline Solids, 2004. 3. P. P. Woskov, S. K. Sundaram, G. Daniel, D. H. Miller, "Millimeter-Wave Measurements of Nuclear Waste Glass Melts", IRMMW-THz 2005 Conference Proceedings, Williamsburg, VA, Sept. 19-23, 2005 4. P. P. Woskov, S. K. Sundaram, G. Daniel, J. S. Machuzak, P. Thomas, "Millimeter-Wave Monitoring of Nuclear Waste

Glass Melts, An Overview", Environmental Issues and Waste Management Technologies VII, (Ceramic Transactions, Volume 132) pp. 189-201, 2002 (best paper award American Ceramic Society). 5. "Millimeter-Wave Radiometer Measurement of Emissivity and Temperature by Thermal Return Reflecti" P. Woskov, K. Hadidi, S. K. Sundaram, and W. E. Daniel, Jr., International Conference on Infrared and Millimeter-Waves, IEEE 02EX561, 211-212, San Diego, Sept. 22-26, 2002. 6. P. P. Woskov, S. K. Sundaram, William E. Daniel, Jr., "Waste Glass Melter Process Monitoring with Millimeter Waves", Spectrum 2002, 9th Biennial Conference on Nuclear and Hazardous Waste Management, American Nuclear Society, 5 pages, Reno, NV, August 4-8, 2002. 7. Paul P. Woskov, K. Hadidi, L. Bromberg, S. K. Sundaram and L. A. Rodgers, Gene Daniel and Don Miller, "Glass Melt Emissivity, Viscosity, and Foaming Monitoring with Millimeter-Waves", 226h American Chemical Society Meeting CD, Division of Nuclear Chemistry & Technology, 81, New York, Sept. 7-11, 2003. 8. S. K. Sundaram, P.P. Woskov, J.S. Machuzak, and W.E. Daniel, Jr., "Cold Cap Monitoring using Millimeter Wave Technology", in Environmental Issues and Waste Management Technologies VII, Editors: G. L. Smith, S. K. Sundaram, and D. R. Spearing (Ceramic Transactions, Volume 132) pp. 203-213, 2002. 9. P. P. Woskov, S. K. Sundaram, W. E. Daniel, Jr., D. Miller, J. Harden, "Millimeter-wave measurements at 137 GHz of DWPF black frit glass flow and salt layer pooling in a pilot scale melt" 227th American Chemical Society Meeting CD, Division of Environmental Chemistry, 63, [psfc.mit.edu/library/04JA001](http://psfc.mit.edu/library/04JA001), March 28-April1, 2004.